Modified spin-wave study of random antiferromagnetic-ferromagnetic spin chains

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We study the thermodynamics of one-dimensional quantum spin-1/2 Heisenberg ferromagnetic system with random antiferromagnetic impurity bonds. In the dilute impurity limit, we generalize the modified spin-wave theory for random spin chains, where local chemical potentials for spin-waves in ferromagnetic spin segments are introduced to ensure zero magnetization at finite temperature. This approach successfully describes the crossover from behavior of pure one-dimensional ferromagnet at high temperatures to a distinct Curie behavior due to randomness at low temperatures. We discuss the effects of impurity bond strength and concentration on the crossover and low temperature behavior.

I. INTRODUCTION

The renormalization group (RG) approach of Wilson[1] provided a framework in which to understand the behavior of various phases of thermodynamic systems in terms of trajectories and RG flows in Hamiltonian space. In addition, it provided a scheme, to calculate, in principle, the behavior in the vicinity of critical and fixed points, by linearizing the flow equations. The method has proved to be powerful in providing an understanding of uniform (translationally invariant) systems, an exact implementation of the Wilson method in systems with quenched random disorder has been relatively restricted. Thus, for example, while the method has been successfully applied to the Anderson localization problem in $(2+\epsilon)$ dimensions,[2] generalization to include electron-electron interactions[3] have had limited success, because of the complications such as the existence of local moments[4,5] which are left out in such long-wavelength approaches to the problem. Another class of problems where a large amount of work has been done[6] is based on the random antiferromagnetic (AF) spin-1/2 chain in one dimension, where it has been shown[7] that a perturbative real space renormalization group (RSRG) scheme becomes asymptotically exact, as has been explicitly verified in a number of cases, such as the random XY chain, where it can be mapped on to a model of free fermions.[8]

Another issue where less attention has been given, is the range of validity of the asymptotic results of the linearized RG equations. In most models of uniform systems, a few iterations of the RG equations leads to the vicinity of the appropriate fixed point. Consequently, a study of the RG trajectory from the region in the vicinity of an critical fixed point to a stable fixed point, usually yields a relatively quick crossover behavior. However, in a study of a three dimensional model related to the 1D random AF chain,[9] it was found that the system did not settle down to a fixed-point-like behavior over several orders of magnitude in temperature; instead there was a slow, logarithmic in temperature, evolution in the behavior. Indeed, it has been claimed recently[10] that the true low temperature behavior of the model is likely different. Unfortunately, such a change cannot be experimentally investigated, because smaller terms in the full Hamiltonian (such as hyperfine coupling to nuclear spins), and dipolar couplings, which have been neglected in the idealized model, will modify the behavior at such (exponentially low) temperatures. Nevertheless, it raises the issue that in models with quenched disorder, the true asymptotic fixed point behavior, may not be practically reached, at least in some cases, making "relevant" (in the RG sense) irrelevant in any practical sense. It is therefore of considerable interest to examine models with quenched disorder where crossovers between short length scale and long length scale behavior can be studied in some detail.

One model, which has received extensive attention in recent years, is the random antiferromagnetic-ferromagnetic (AFM-FM) spin chain.[11,12,13,14,15,16,17,18] The study of such systems has been motivated by the discovery of novel one-dimensional (1D) random spin systems. One example of such materials is $Sr_3CuPt_{1-x}Ir_xO_6$,[19] alloy of the pure compounds Sr_3CuPtO_6 (antiferromagnet) and Sr_3CuIrO_6 (ferromagnet). Such a system is interesting due to the interplay between quantum fluctuations and disorder; the latter can not be treated as a perturbation, as it is often found to change the low-energy spectra dramatically or even to destabilize the pure phases. Theoretical and numerical methods used to study random antiferromagnetic-ferromagnetic spin chains have been largely inherited from the study of random antiferromagnetic (AFM) chains (with AFM coupling only).[7,20,21] In particular, the real-space renormalization group (RSRG) approach[12,14] revealed a Curie-like temperature dependence in the susceptibility of a random AFM-FM spin-1/2 chain at low temperatures. In the RSRG picture, the Curie behavior occurs because spins correlate, due to the existence of ferromagnetic couplings, to form clusters whose average size and effective spin grow in a random walk fashion at low temperatures. Therefore, the random AFM-FM spin-1/2 chain belongs to a different

universality class from that of the random AFM spin-1/2 chain, whose ground state is known as a random singlet phase, in which singlets can be formed over large distance. The RSRG results have been supported by various other numerical techniques, such as quantum Monte Carlo (QMC) simulation[16,17,32] and density matrix renormalization group (DMRG) method.[15,18]

In the RSRG method, one decimates the spins that are coupled by the strongest coupling in the system, and renormalizes the couplings among the remaining spins perturbatively. In this procedure new effective couplings and effective spins may also be generated. This procedure, which in general lowers the overall energy scale of the problem at hand, is repeated so that excitations at lower and lower energy scales are probed. Its quantitative accuracy relies on the presence of *strong* randomness, as in the case of strong randomness the strength of the strongest and typical couplings are well separated, ensuring the accuracy of a perturbative calculation. In real systems, however, the randomness can be very weak, especially when it is not introduced intentionally. In this case the RSRG is *not* expected to be quantitatively reliable in its early stages. It has been argued[7,14] that the strength of disorder grows as the energy scale is lowered, thus in the low-energy (or equivalently, low-temperature) limit, the RSRG results become correct or even asymptotically exact. Nevertheless in this case one expects the thermodynamic properties of the system to be dominated by the physics of the pure chains at high temperature, and crossover to the randomness dominated regime (where the RSRG results apply) at very low temperatures.

In the present work we study the thermodynamic properties of the random AFM-FM chain, with mostly uniform FM coupling and a small concentration of (impurity) AFM bonds, using the modified spin-wave method. We also compare the results of modified spin-wave method with those of exact diagonalization in small systems to demonstrate the accuracy of the former. Our motivation comes from the following considerations. First of all, as discussed above, when the concentration of the impurity bonds is small, the randomness is rather weak and the RSRG results are not reliable at high temperatures. The modified spin-wave method allows us to treat the high and low temperature regimes on equal footing, and in particular, address the crossover between these two regimes. This makes it possible to compare theory with experiments for the entire temperature range, when the randomness is weak. Secondly, the modified spin-wave method has so far been applied to study pure spin models only. While very successful in those cases, it has not yet been used to study random spin systems. By working out a number of technical issues that one faces when applying it to random spin problems, and demonstrate its accuracy in the present problem, we lay the ground for the application of this powerful method to other random spin problems. Thirdly, as discussed in the opening paragraphs, it is of general interest to study models where the short distance (high temperature) and long distance (low temperature) behavior are controlled by different fixed points in the renormalization group sense, and address the crossover between the two limiting cases quantitatively; our model is such an example, and we hope our study will stimulate future research on this important issue.

Our results are summarized as follows. We find that the spin susceptibility of our system follows that of a pure ferromagnetic chain at high temperatures, and crosses over to $1/T$ dependence at low temperatures predicted by the RSRG, with a coefficient that agrees with the RSRG result essentially exactly. We also find that the crossover temperature depends mainly on the impurity bond *concentration*, while the width of the crossover depends mainly on the impurity bond *strength*; we determine their dependences semiquantitatively. We also demonstrate that the results of the modified spin-wave method is asymptotically exact in certain limiting cases, and quite accurate in the entire temperature range by comparing them with exact diagonalization results in the systems.

The paper is organized as follows. In Sec. II, we describe the model we study, as well as the qualitative physics expected from the model. In Sec. III, we introduce the modified spin-wave theory for the random AFM-FM spin chains. We present the numerical results of the theory in Sec. IV, with emphasis on the effects impurity bond strength, impurity concentration, as well as finite-size effects of our numerical study on the spin susceptibility. Finally, our results are summarized in Sec. V. Readers interested in the comparison of the modified spin-wave theory to exact diagonalization results will find details in Appendix A and B for simple cases like single ferromagnetic segment and two coupled segments.

II. THE QUALITATIVE PICTURE

We consider the following Hamiltonian for a random spin-S chain

$$H = \sum_i J_i \mathbf{S}_i \cdot \mathbf{S}_{i+1}, \tag{1}$$

where the coupling strength J_i is randomly chosen according to the distribution

$$P(J) = (1-p)\delta(J + J_F) + p\delta(J - J_{AF}). \tag{2}$$

J_F and J_{AF} are the coupling strengths of the ferromagnetic and antiferromagnetic bonds, respectively. p is the concentration of the antiferromagnetic impurity bonds. We are primarily interested in the dilute doping regime, *i.e.*, $p \ll 1$, so that we have long ferromagnetic spin segments, separated by antiferromagnetic bonds. The number of spins in a spin segment (N_s) is random, with an exponential distribution

$$P(N_s) = p(1-p)^{N_s-1}. \tag{3}$$

Therefore, the average number of spins per segment is $\langle N_s \rangle = 1/p$. From real space renormalization group studies[9,20], we understand that a strong antiferromagnetic bond tends to lock the two adjacent spins into a singlet (which is inert at temperatures of interest), and the singlet thereby generates a weaker effective AF coupling between the two spins next to the singlet (see Appendix B). Therefore, for simplicity, we consider weak antiferromagnetic coupling ($J_{AF} < J_F$) only without losing generality.

The temperature dependence of magnetic susceptibility χ per spin is qualitatively known. We assume the Zeeman term $H' = -\mu H_z \sum S_i^z$ and set $\mu = k_B = 1$ for simplicity. At high temperatures ($T \gg J_F$), the spins behave independently. Therefore, we expect an ordinary Curie contribution of

$$\chi_{HT} = \frac{S(S+1)}{3T}, \tag{4}$$

from each spin. When $T \leq J_F$, the spins within each ferromagnetic segment become correlated. Takahashi[25] obtained the following low temperature expansion for spin-1/2 ferromagnetic chain (of infinite length)

$$\chi_{FM} = \frac{1}{4}\left[\frac{0.1667 J_F}{T^2} + \frac{0.581 J_F^{1/2}}{T^{3/2}} + \frac{0.68}{T} + O\left(T^{1/2}\right)\right] \tag{5}$$

using the Bethe-ansatz integral equations. The factor of 1/4 is added here due to an extra factor of 2 in the Zeeman energy terms in the original paper. Therefore, susceptibility of each segment rises as temperature decreases, crossing over from $1/T$ to $1/T^2$. In the case of $J_{AF} = 0$, χ crosses over back to Curie behavior at lower temperatures, when each segment of finite length acts like a block spin with frozen internal excitations. When T drops below the typical spin-wave excitation gap of a typical segment

$$\Delta_{SW} \sim J_F S \left(\frac{2\pi}{\bar{N}_s}\right)^2, \tag{6}$$

we expect

$$\chi_{BS} \equiv \frac{c_{BS}}{T} = \frac{1}{N}\sum_s \frac{N_s S(N_s S+1)}{3T}. \tag{7}$$

Here $\bar{N}_s$ is the typical length of a ferromagnetic segment. In the thermodynamic limit, we obtain the Curie constant for locked block spins to be

$$c_{BS} = \frac{S^2}{3}\frac{2-p}{p} + \frac{S}{3}, \tag{8}$$

using segment-size distribution in Eq. 3. At $p = 0.1$, we obtain $c_{BS} = 7/4$ for $S = 1/2$. c_{BS} decreases with increasing impurity concentration p and is always (unless $p = 1$, *i.e.* in the pure antiferromagnetic case) greater than the high temperature Curie constant (in Eq. 4)

$$c_{HT} = \frac{S(S+1)}{3} = \frac{1}{4}, \text{ for } S = \frac{1}{2}, \tag{9}$$

expected at high temperatures.

When the end spins of neighboring segments are coupled with weak antiferromagnetic coupling J_{AF}, the block spins (originating from the ferromagnetic segments) interact with weak effective couplings. For two segments with N_1 and N_2 spins, we obtain the effective coupling

$$J' = \frac{J_{AF}}{N_1 N_2}, \tag{10}$$

by projecting the coupling to the block-spin space using Wigner-Eckart theorem. Not surprisingly, long segments are weakly coupled. In the RSRG approach,[13] one starts with two strongly coupled segments and decimates the segments to a $S' = |N_1 - N_2|S$ spin, and renormalize its couplings with neighboring segments. The spin-wave excitations within each segment, considered in our modified spin-wave approach, are in general neglected in the RSRG approach, which is, therefore, valid only at low enough temperatures. By including these spin-wave excitations, we can not only demonstrate that the Curie susceptibility (predicted by RSRG) in a AFM-FM spin-1/2 chain at low temperatures is indeed the result of correlated ferromagnetic segments, but estimate the temperature scale at which the low temperature Curie behavior occurs as well. Since the renormalized couplings may become ferromagnetic, clusters start to grow in a random walk fashion as segments couple to neighboring segments as we lower the temperature. As in a typical random walk problem, we expect the average spin $\bar{S}_l$ and the average size $\bar{l}$ of such clusters of spins satisfy

$$\bar{S}_l \sim \bar{l}^{-1/2}, \tag{11}$$

so that as far as magnetic susceptibility per spin χ is concerned, the scaling behavior of $\bar{S}_l^2$ and $\bar{l}$ cancels, leading to

$$\chi_{LT} = \frac{c_{LT}}{T} = \frac{1}{3T}\frac{\bar{S}_l^2}{\bar{l}} \sim \frac{1}{T}. \tag{12}$$

By statistical analysis,[14] one finds the low-temperature Curie constant to be

$$c_{LT} = \frac{S^2}{3}\frac{1-p}{p}, \tag{13}$$

in the large cluster-size limit. This result coincides with the classical result for the susceptibility per spin at low temperatures.[11,19] Compared with Eq. 8, we find that antiferromagnetic impurity bonds reduce the low-temperature Curie constant by

$$\Delta c = c_{BS} - c_{LT} = \frac{S^2}{3p} + \frac{S}{3}. \tag{14}$$

On the other hand, for small p (or low concentration of AF bonds), we have $c_{LT} > c_{HT}$; this simply reflects the fact that the dominant interaction in the system is ferromagnetic, which enhances spin susceptibility. Thus in the cases we are interested in, we have

$$c_{HT} < c_{LT} < c_{BS}. \tag{15}$$

We should note here that unless J_{AF} is very small, there may not be a temperature range in which $\chi \approx \chi_{BS} = c_{BS}/T$; this is because the neighboring segments can start developing correlations due to J_{AF} *before* the intrasegment spin-wave excitations are frozen out by temperature. We will actually study mostly this case in this work, and focus mainly on the crossover from the pure ferromagnetic chain behavior directly to the asymptotic low temperature behavior with c_{LT} as the Curie coefficient. We do this partly because in real systems J_{AF} is often comparable to J_F, and partly for the sake of simplicity. Our method, however, is capable of handling very small J_{AF} and the regime with c_{BS} being the Curie coefficient.

III. THE MODIFIED SPIN-WAVE APPROACH

The conventional spin-wave theory gives the exact spin-wave spectra of Heisenberg ferromagnets at $T = 0$. At finite temperature however, the theory leads to difficulty in one-dimensional quantum ferromagnets, the number of spin waves diverges when external magnetic field goes to zero. This difficulty has its root in the Mermin-Wagner theorem, which dictates that there cannot be long-range order in 1D at finite temperature. By imposing a constraint that the total magnetization be zero (thus fulfilling the Mermin-Wagner theorem), Takahashi[26] introduced a chemical potential (equivalence to a uniform magnetic field) for the spin waves and successfully obtained the low-temperature properties of the ferromagnets in one dimension, as well as in two dimensions. The beauty of this modified spin-wave theory is that it gives correct asymptotic thermodynamic behavior at both large and zero temperature limit, as well as fairly accurate crossover behavior at intermediate temperatures. The modified spin-wave theory have further succeeded in two-dimensional antiferromagnets[27,28] and one-dimensional ferrimagnets.[29] For one-dimensional antiferromagnets, the modified spin-wave theory predicted a gap in the spin-wave spectrum,[30] consistent with the Haldane gap in integer-spin chains.[31]

In this paper, we extend its applicability to ferromagnetic spin chains with dilute antiferromagnetic impurities. In the dilute impurity limit, a spin chain consists of ferromagnetic spin segments, coupled antiferromagnetically. The application of the modified spin-wave theory to the random antiferromagnetic-ferromagnetic spin chains is, therefore, mainly based on our observation that ferromagnetic spin segments can be well described by the modified spin-wave theory (see Appendix A). We found that, for magnetic susceptibility of a finite spin chain, the modified spin-wave theory gives noticeable difference from the exact diagonalization result only at two temperature crossovers. The crossover at higher temperatures describes the switching on of the couplings between individual spins, which is of little interest in the competition between different segments. On the other hand, the maximum error of the modified spin-wave theory is roughly 5%, for a chain of up to 14 spins, near the lower-temperature crossover when the spin chain becomes locked into one block spin. For a finite spin chain, the translational symmetry is broken. Therefore, one, in principle, needs to introduce a local chemical potential (or a local field) for each spin to ensure the magnetic moment of the spin be zero. Fortunately, we can apply a periodic boundary condition to each ferromagnetic spin segment, so that only one chemical potential is needed throughout the segment. We emphasize that the use of periodic boundary condition reduces the computational complexity without changing the essential physics. This is particularly true when we have very dilute impurities so that the average ferromagnetic segment length is large.

For the nearest-neighbor Heisenberg Hamiltonian, Eq. 1, we apply Holstein-Primakoff transformation

$$\begin{cases} S_i^{s+} &= \sqrt{2S - a_i^{s\dagger} a_i^s}\, a_i^s, \\ S_i^{s-} &= a_i^{s\dagger} \sqrt{2S - a_i^{s\dagger} a_i^s}, \\ S_i^{sz} &= S - a_i^{s\dagger} a_i^s. \end{cases} , \tag{16}$$

for segments with odd index s, and

$$\begin{cases} S_i^{s+} &= a_i^{s\dagger} \sqrt{2S - a_i^{s\dagger} a_i^s}, \\ S_i^{s-} &= \sqrt{2S - a_i^{s\dagger} a_i^s}\, a_i^s, \\ S_i^{sz} &= a_i^{s\dagger} a_i^s - S. \end{cases} , \tag{17}$$

for segments with even index s. This way, we introduce a distinct species of boson for each segment. The Hamiltonian, in the linear spin-wave approximation, becomes

$$\begin{aligned} H = E_0 \;+\; & J_F S \sum_{s=1}^{M_s} \sum_{i=1}^{N_s} \left(a_i^{s\dagger} a_i^s + a_{i+1}^{s\dagger} a_{i+1}^s - a_i^{s\dagger} a_{i+1}^s - a_{i+1}^{s\dagger} a_i^s \right) \\ + \; & J_{AF} S \sum_{s=1}^{M_s - 1} \left(a_{N_s-1}^{s\dagger} a_{N_s-1}^s + a_1^{(s+1)\dagger} a_1^{(s+1)} + a_{N_s-1}^{s\dagger} a_1^{(s+1)} + a_1^{(s+1)\dagger} a_{N_s-1}^s \right), \end{aligned} \tag{18}$$

where M_s is the number of ferromagnetic segments, and N_s the number of spins in the s-th segment. The total number of spins is, therefore, $N = \sum_{s=1}^{M_s} N_s$. Note $(N_s + 1) \equiv 0$, reflecting the periodic boundary condition imposed on each segment.

In the linear spin-wave approach, the quadratic Hamiltonian is soluble by a generalized Bogoliubov transformation for bosons, a generalization of Ref. 24. The procedure can be written compactly in a matrix format. Denote the original boson operators by a vector

$$x = (a_1^1, a_2^1, \cdots, a_{N_1}^1, a_1^{2\dagger}, a_2^{2\dagger}, \cdots, a_{N_2}^{2\dagger}, \cdots)^T. \tag{19}$$

The Hamiltonian can be written as

$$H = const. + x^\dagger \mathcal{H} x, \tag{20}$$

where

$$\mathcal{H} = J_F S \begin{pmatrix} 2 & -1 & & & -1 & & & & & & & \\ -1 & 2 & \ddots & & & & & & & & & \\ & \ddots & \ddots & & -1 & & & & & & & \\ -1 & & -1 & 2+J_{AF}/J_F & J_{AF}/J_F & & & & & & & \\ & & & J_{AF}/J_F & 2+J_{AF}/J_F & -1 & & & -1 & & & \\ & & & & & -1 & & 2 & \ddots & & & \\ & & & & & & \ddots & \ddots & & -1 & & \\ & & & & & -1 & & & -1 & 2+J_{AF}/J_F & J_{AF}/J_F & \\ & & & & & & & & & J_{AF}/J_F & 2+J_{AF}/J_F & \ddots \\ & & & & & & & & & & \ddots & \ddots \end{pmatrix}, \tag{21}$$

is a $N \times N$ matrix. The Hamiltonian matrix is basically a 3-diagonal matrix, with extra (-1)'s at the corners of each block (of a spin segment). The rest of the matrix elements (left unspecified in Eq. 21) are zeros. Introduce the generalized Bogoliubov transformation:

$$x = V\gamma, \tag{22}$$

where V is a $N \times N$ matrix and γ a vector of the set of boson operators (α's)

$$\gamma = (\alpha_1^1, \alpha_2^1, \cdots, \alpha_{N_1}^1, \alpha_1^{2\dagger}, \alpha_2^{2\dagger}, \cdots, \alpha_{N_2}^{2\dagger}, \cdots)^T, \tag{23}$$

that diagonalizes $\mathcal{H}$, *i.e.*,

$$V^T \mathcal{H} V = E, \tag{24}$$

in the compact matrix format, where E is a diagonal matrix,

$$E_{N\times N} = diag(\varepsilon_1^1, \varepsilon_2^1, \cdots, \varepsilon_{N_1}^1, \varepsilon_1^{2\dagger}, \varepsilon_2^{2\dagger}, \cdots, \varepsilon_{N_2}^{2\dagger}, \cdots, \varepsilon_{N_{M_s}}^{M_s\dagger}) \tag{25}$$

with its diagonal elements being the energies of the corresponding bosons after the Bogoliubov transformation. The boson commutation relation requires

$$VPV^T = P, \tag{26}$$

where P is a generalized *para unit matrix* (inheriting the notation from Ref. 24) of the form

$$P_{N\times N} = diag(\overbrace{1, 1, \cdots, 1}^{N_1}, \overbrace{-1, -1, \cdots, -1}^{N_2}, \cdots) \tag{27}$$

For fermions, on the other hand, the fermionic commutation relations lead to the orthonormalization condition, which can assume the same equation as Eq. 26, except that P becomes an $N \times N$ unit matrix. We need to find out a solution V for bosons, which simultaneously satisfies Eq. 24 and 26; this can be done with the help of generalized matrix diagonalization. Unlike fermionic diagonalization, the existence of such a solution is not guaranteed, unless spin segments are decoupled, *i.e.* $J_{AF} = 0$. Such unfortunate situations are, in general, associated with modes of zero energy, which then leads to divergent number of bosons (or divergent magnetization). In the case of a single ferromagnetic chain, Takahashi[26] introduced a chemical potential (or a magnetic field) to overcome the difficulty. In this case, we need to introduce to the Hamiltonian a set of M_s local chemical potentials, one for each segment, to find a solution. These chemical potentials are chosen such that the magnetization of each spin segment (equivalently, of each spin with periodic boundary condition) is zero. These constraints, which have the physical significance of restoring the rotational symmetry, ensure that the number of bosons be finite at finite temperatures. The thermodynamic quantities can be calculated from the excitation energies ε's and the transformation matrix V. In particular, magnetic susceptibility per site χ can be written as

$$\chi = \frac{1}{3TN} \sum_{s=1}^{M_s} \sum_{i=1}^{N_s} \tilde{n}_i^s \left(\tilde{n}_i^s + 1\right), \tag{28}$$

where $\tilde{n}_i^s$ is the occupation number for the boson correspond to operator α_i^s and excitation energy ε_i^s,

$$\tilde{n}_i^s = \langle \alpha_i^{s\dagger} \alpha_i^s \rangle = \frac{1}{e^{\varepsilon_i^s/T} - 1}. \tag{29}$$

IV. RESULTS FOR MAGNETIC SUSCEPTIBILITY

Figure 1 shows the static magnetic susceptibility χ per spin of a 600-spin ferromagnetic chain, with 59 antiferromagnetic impurity bonds, which divide the chain into 60 ferromagnetic segments, *i.e.*, $p = 0.1$. The impurity bonds ($J_{AF} = 0.5$) are put randomly with the constraint that the total magnetization of the corresponding classical ground state is zero. This guarantees that χ drops to zero at zero temperature. The susceptibility curve shows three temperature regimes. For $T/J_F > 3$, result can be fit to a high-temperature Curie law (Eq. 4), suggesting that spins behave independently. For $0.2 < T/J_F < 3$, χ rises with decreasing temperature, following Eq. 5. This implies that spins start to correlate, forming independent ferromagnetic segments. The antiferromagnetic coupling is still too weak to affect thermodynamics of the random spin chain at these temperatures. Below $T = 0.2J_F$, χ is in good agreement with

$$\chi = \frac{c_{LT}}{T}, \qquad c_{LT} = \frac{3}{4}, \tag{30}$$

the low-temperature Curie behavior expected by the RSRG approach (Eq. 13 for $p = 0.1$). We point out that the Curie constant of independent block spins (Eq. 8) is expected to be $c_{BS} = 7/4$ for $p = 0.1$. Therefore, the low-temperature Curie constant is indeed reduced by antiferromagnetic couplings between ferromagnetic segments. We note that χ deviates from Eq. 30 below $T = 0.02J_F$. We believe this is an artifact due to finite size (we have only 600 spins), since there are no more segments to decimate. χ bends down as we specially require that χ go to zero at zero temperature. Since the number of iterations required to find a solution for the generalized Bogoliubov transformation grows significantly at low temperatures (nearly 1000 iterations at $T = 0.005$), we were unable to approach temperatures much lower than $T = 0.01$ or to calculate many samples on our regular workstations. It is worth pointing out that we can identify the crossover temperature from the ferromagnetic spin-chain physics to random spin-chain physics as the cross point of Eq. 30 and 5. In the dilute doping limit (small p), we have

$$T_x = J_F \frac{p}{1-p}\left[0.50 + 3.04\sqrt{\frac{p}{1-p}} + O\left(\frac{p}{1-p}\right)\right] \tag{31}$$

For $p = 0.1$, $T_x = 0.168J_F$.

Figure 2 shows the susceptibility per spin χ of spin chains of 20, 30, and 40 ferromagnetic segments (*i.e.*, with 19, 29, and 39 antiferromagnetic impurity bonds, respectively). The average ferromagnetic segment length is fixed at 8 spins ($p = 1/8$), so that the lengths of the corresponding spin chains are 160, 240, and 320, respectively. The antiferromagnetic coupling J_{AF} is chosen again to be 0.5. χ is averaged over 10-20 random realizations depending on size. For $0.2 < T/J_F < 1$, χ roughly follows the low-temperature (compared with J_F) expansion of a single ferromagnetic chain (Eq. 5). From now on, we neglect the thermodynamics at $T > J_F$ (although it is capable of being calculated within our theory), since it is trivial and not the interest of this paper. Below the crossover temperature $T_x \sim 0.2$, χ deviates from the pure chain susceptibility, and obeys a Curie law. The range of the Curie susceptibility extends to lower and lower temperature when we have more segments (from 20 to 40 in Fig. 2), therefore longer chain, for a fixed impurity concentration. At $N_s = 40$, this range extends beyond one decade for $p = 1/8$. We point out that the Curie constant obtained from Fig. 2 ($c = 0.67$) is roughly 15% larger than predicted by Eq. 13. This should be viewed as a mixture of both the RSRG-predicted behavior and the ferromagnetic-chain physics. The reason will become clear later after we explore the impurity concentration dependence of the Curie behavior.

The antiferromagnetic coupling strength J_{AF} determines the temperature scale at which the ferromagnetic segments couple to their nearest neighbors. The smaller J_{AF} is, the lower the temperature at which RSRG results are valid becomes, leading therefore to an increasingly wider crossover from the physics of a pure ferromagnetic chain. Figure 3 shows the sample averaged magnetic susceptibility per spin χ for $J_{AF} = 0.3$ and 0.5 for spin chains of 30 ferromagnetic segments. The average number of spins in each segment is 12 spins ($p = 1/12$). The dashed line corresponds to a Curie constant $c = 11/12$, expected by Eq. 13 for $p = 1/12$. Although the system size (360 spins) is probably not large enough, one can nevertheless identify the Curie behavior for $J_{AF} = 0.5$ around $T = 0.05$ and for $J_{AF} = 0.3$ around $T = 0.02$. Note that χ for $J_{AF} = 0.3$ is greater than that for $J_{AF} = 0.5$ at low temperatures. This is expected because decimated spin segments, in the RSRG language, contribute less to the magnetic susceptibility than the sum of original segments, and because the decimation happens at lower temperatures for smaller J_{AF}. For very small J_{AF}, we expect the system crosses over from a ferromagnetic regime at high temperatures, first, to an (almost) independent block-spin regime at lower temperatures, characterized by a Curie constant as in Eq. 8. There is another crossover, at very low temperatures (dependent on J_{AF}), from the independent block-spin regime to the RSRG regime, characterized by a smaller Curie constant as in Eq. 13.

We have also studied the dependence of the magnetic susceptibility on the impurity concentration, or equivalently, average number of spins in each segment. Qualitatively, the larger number of spins within each segment increases the

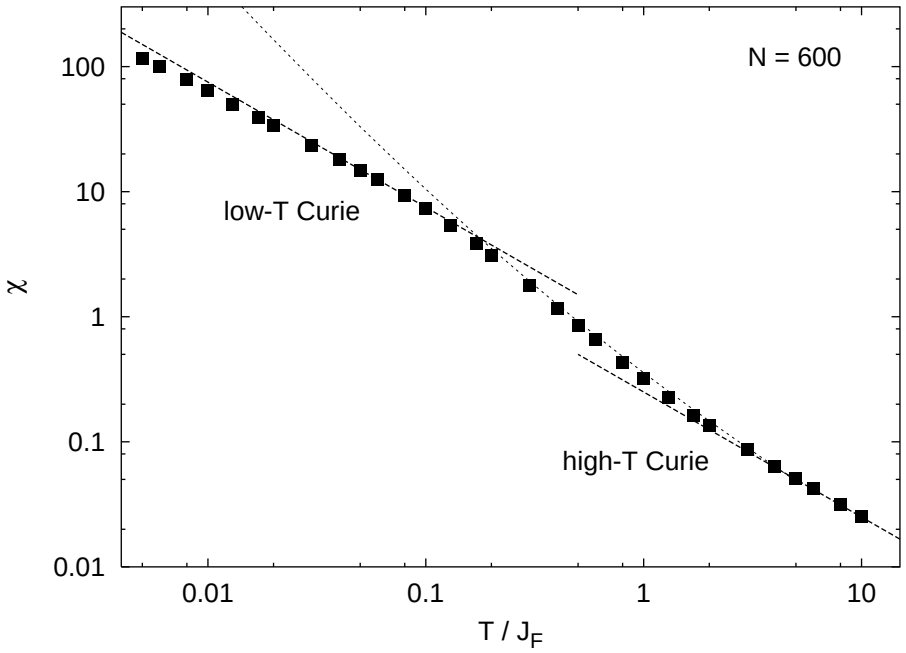

FIG. 1: Static magnetic susceptibility per spin (χ) of a single random Heisenberg spin chain of 600 spins. The spins are coupled with their nearest neighbors by ferromagnetic couplings (J_F), with 59 antiferromagnetic impurity couplings ($J_{AF} = 0.5J_F$), so that the spin chain can be viewed as 60 ferromagnetic segments of average length 10 (spins). On the log-log scale, the two dashed lines with slope unity are exact results of the ordinary Curie susceptibility of independent spins at high temperatures (Eq. 4) and the low-temperature Curie susceptibility expected by the RSRG approach (Eq. 13). The dotted line is the low temperature expansion of the magnetic susceptibility (Eq. 5) of a pure spin-1/2 ferromagnetic chain (of infinite length), which is proportional to $1/T^2$ in the low temperature limit.

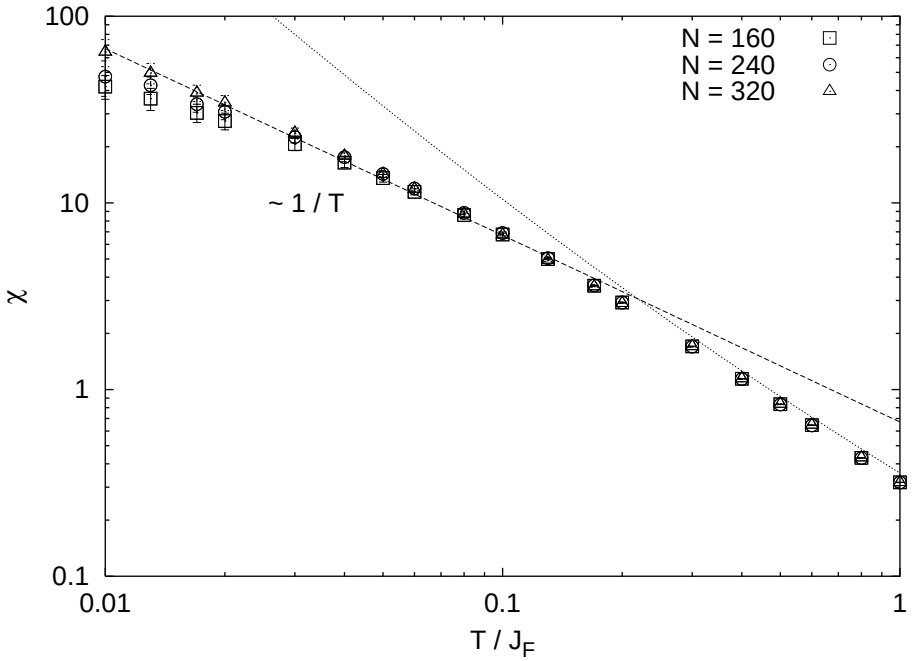

FIG. 2: Sample averaged magnetic susceptibility per spin for spin chain of 160, 240, 320 spins, which consists of 20, 30, 40 ferromagnetic segments, respectively, coupled by impurity antiferromagnetic bonds with $J_{AF} = 0.5J_F$. The dotted line is the low temperature expansion of the magnetic susceptibility (Eq. 5) of a spin-1/2 ferromagnetic chain (of infinite length). The dashed line is a fit to the apparent (see text for detail) low-temperature Curie susceptibility.

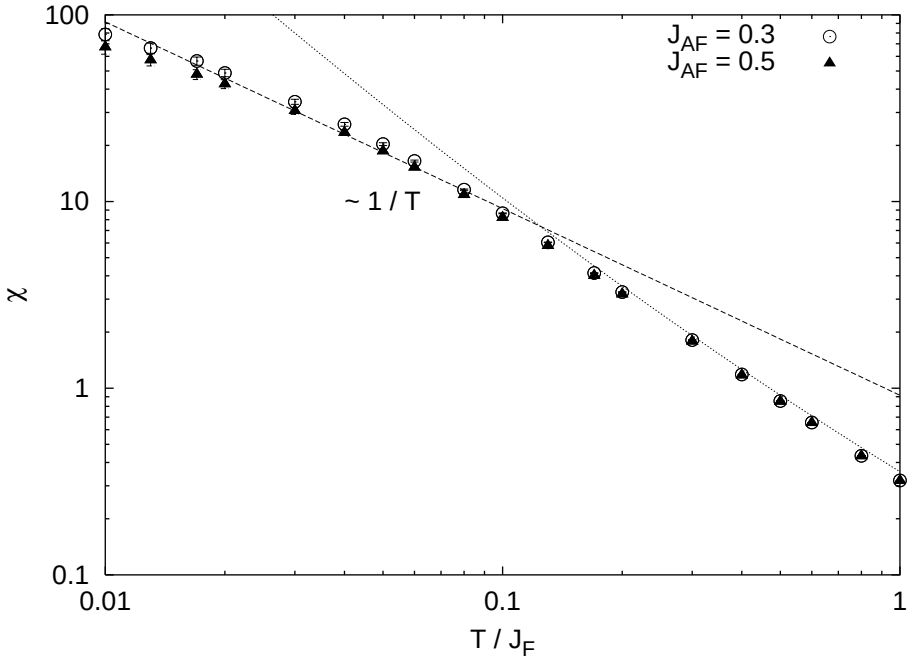

FIG. 3: Sample averaged magnetic susceptibility per spin for spin chains of 360 spins in 30 ferromagnetic segments, coupled by antiferromagnetic bonds with $J_{AF} = 0.3J_F$ and $0.5J_F$. The data is expected to agree with the low-temperature Curie susceptibility (the dashed line) in the RSRG method. The temperature range of the agreement is expected to be higher for stronger antiferromagnetic impurity bonds ($J_{AF} = 0.5J_F$). The dotted line is the low temperature expansion of the magnetic susceptibility (Eq. 5) of a spin-1/2 ferromagnetic chain (of infinite length).

magnetic susceptibility per spin, since $\chi \sim N_s^2$ for a ferromagnetic N_s-spin segment (for large N_s). As a consequence, the Curie constant is expected to increases with the average number of spins in each segment, or more precisely, as in Eq 13. This trend is shown in Fig. 4, where we plot the averaged susceptibility per spin for 30-segment spin chains, with 8, 12, and 16 spins per segment on average, respectively. The three dashed lines are the low-T Curie behavior the RSRG approach expected (Eq. 13). As is pointed out in earlier paragraph, our data is in significant disagreement with Eq. 13 for $p = 1/8$. For $p = 1/16$, one finds that χ follows Eq. 13 from $T = 0.015J_F$ to 0.04, the regime antiferromagnetic impurity bonds are dominating. The Curie regime shifts to $0.02 < T/J_F < 0.08$ for $p = 1/12$. This is consistent with Eq. 10 that longer segments have relatively weaker effective intersegment coupling. The trend suggests that for $p = 1/8$ the Curie regime would occur above $T > 0.1J_F$, which coincides with the regime where intersegment coupling is important. Therefore, we believe the discrepancy between our data and Eq. 13 reveals a collective contribution from the ferromagnetism with each segment and the antiferromagnetism between segments. This demonstrates that inappropriate parameters can kill (or lead to the wrong) RSRG physics. We note that the crossover regime (deviating from both Eq. 4 and the $1/T$-law) becomes wider for smaller p. This is again consistent with the translation between low concentration and weak effective intersegment coupling, which, we have demonstrated, leads to wider crossover regime.

V. CONCLUSION

In the study of the random AFM-FM (nearly ferromagnetic) spin-1/2 chain, the modified spin-wave theory allows us to obtain the thermodynamic properties over a broad temperature range (on logarithmic scale). We have studied systems as large as 600 spins, with up to 60 ferromagnetic segments, for temperature ranges of over three orders of magnitude. At very high temperatures, the spins are independent; correlation starts to develop within each ferromagnetic segments as temperature drops, giving rise to characteristics of a pure ferromagnetic chain. At low temperatures, the observation of the Curie-like behavior reveals the regime where ferromagnetic segments are correlated into clusters, whose size grows with decreasing temperature. Therefore, we can study, fairly systematically, in the modified spin-wave theory the crossover between the high-temperature Curie behavior (independent spins) and low-temperature Curie behavior (correlated spin segments).

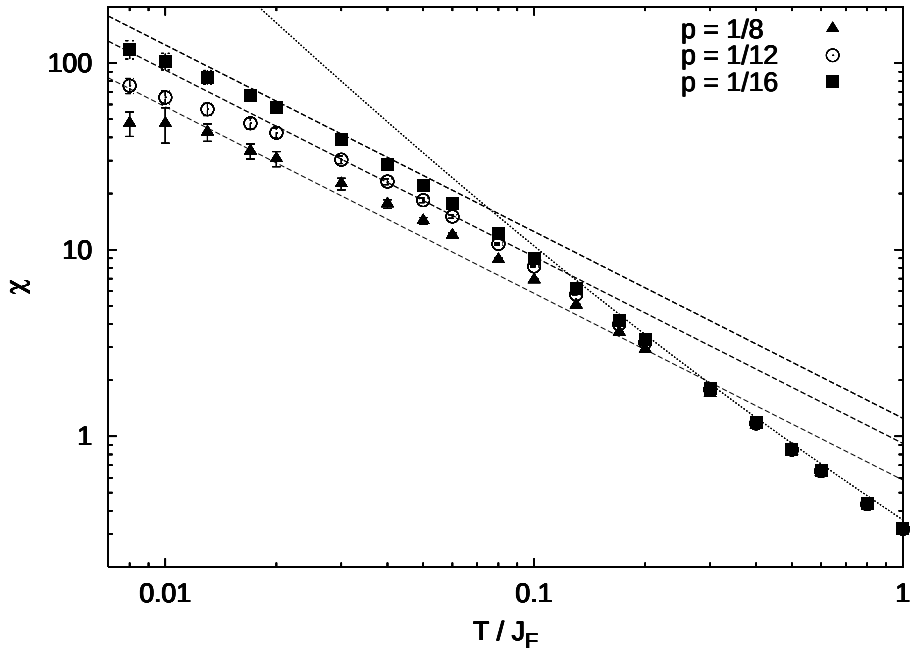

FIG. 4: Sample averaged magnetic susceptibility per spin for spin chains of 240, 360, and 480 spins, each containing 30 ferromagnetic segments, coupled by antiferromagnetic bonds with $J_{AF} = 0.5J_F$. The dotted line is the low temperature expansion of the magnetic susceptibility (Eq. 5) of a spin-1/2 ferromagnetic chain (of infinite length). The dashed lines are the exact results of the low-temperature Curie susceptibility expected by the RSRG approach for the three impurity concentrations.

System size is important in observing the low-temperature Curie behavior, since it determines whether the RSRG scaling regime can be reached. Our study has found that the modified spin-wave theory gives results in accordance with the RSRG argument for chains of several hundred spins (after averaging over random bond distribution). The study of the effects of impurity bond strength shows that the low-temperature Curie regime can be consistently pushed to lower temperatures by decreasing the antiferromagnetic impurity bond strength.

In the dilute doping regime, we have studied the effects of impurity concentration on the low-temperature thermodynamics. The results are again in fairly good agreement with the RSRG prediction. The exception happens at higher concentration ($p = 1/8$), where we believe the intrasegment ferromagnetic coupling and the intersegment antiferromagnetic coupling take actions at about the same temperature regime, leading to a mixed behavior which appears to look like a different Curie behavior. Further explorations using other methods would be very useful to confirm our explanation.

In our study, both weak impurity bonds and low impurity concentration give rise to a wide crossover from the ferromagnetic spin-chain regime to the random spin-chain regime. Although the modified spin-wave theory limits us to only a portion of the whole random spin-chain space, we believe such wide crossover can be generic and it can prevent the real RSRG behavior from being observed at reasonably low temperatures. In fact, the RSRG study of random spin chains[14] has pointed out the possibility of having a crossover region of more than five orders of magnitude before the true RSRG scaling regime can be reached. A quantum Monte Carlo study of the random AFM-FM spin-1/2 chain[32] has confirmed that a region of weakly interacting spin segments can exist, whose crossover to the low-temperature scaling regime is marked by a (somewhat small) peak in the specific heat.

The main difficulty of directly comparing our results with experiments seem to be that quasi-1D Heisenberg ferromagnetic systems are rare since there usually exist interchain couplings which lead to three-dimensional (3D) ordering at low temperatures. For Sr_3CuIrO_6, long-range (presumably 3D) order develops at 20.1 K without signature of 1D magnetism.[22] Recently, however, effects of magnetic impurities on the quasi-1D ferromagnetic spin chain have been reported for organic radical alloy, $(p\text{-CDpOV})_{1-x}(p\text{-BDpOV})_x$,[23] whose interchain-intrachain coupling ratio is as small as $J'/J = 3.7 \times 10^{-3}$. This may open a door where the low-temperature Curie susceptibility of the RSRG origin can be observed in ferromagnetic bond dominated random spin chains.

VI. ACKNOWLEDGMENTS

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APPENDIX A: MODIFIED SPIN-WAVE THEORY FOR FERROMAGNETIC SPIN SEGMENTS

In Takahashi's original paper[26], the modified spin-wave theory is aimed at ferromagnetic spin chains with infinite length. Nevertheless, the theory can be easily applied to a finite-size ferromagnetic spin chain with periodic boundary conditions. This only changes the continuous spin-wave spectrum to a discrete spectrum. Therefore, for an N-spin ferromagnetic chain, we can write the self-consistent equations for the single (global) chemical potential μ:

$$\begin{aligned} NS &= \sum_k \tilde{n}_k, \\ \tilde{n}_k &= \frac{1}{e^{[\epsilon(k)-\mu]/T}-1}, \end{aligned} \tag{A1}$$

where the discrete spin-wave energy spectrum is

$$\epsilon(k) = 2JS(1-\cos ka), \qquad k = \frac{2\pi i}{Na},\ i = 0, 1, ..., N-1. \tag{A2}$$

The magnetic susceptibility per spin can be expressed as

$$\chi = \frac{1}{3TN}\sum_k \tilde{n}_k(\tilde{n}_k+1). \tag{A3}$$

Analytical results, which turn out to be exact, can be obtained for both high and low temperature limit. In the high temperature limit, $\tilde{n}_k = S$ is a constant. Self-consistent Eqs. A1 give

$$\mu = T\ln\left(1+S^{-1}\right). \tag{A4}$$

Therefore, we have

$$\chi = \frac{S(S+1)}{3T}, \tag{A5}$$

which is the exact result for independent spins. In the low temperature limit, only $k=0$ mode can be excited, *i.e.* $\tilde{n}_k = NS\delta_{k,0}$. Self-consistent Eqs. A1 give

$$\mu = T\ln\left(1+N^{-1}S^{-1}\right). \tag{A6}$$

Therefore, we have

$$\chi = \frac{S(NS+1)}{3T}, \tag{A7}$$

which is the exact result for the ferromagnetic ground state with maximum spin.

Figure 5 shows χ for ferromagnetic spin-1/2 chains of finite length $N = 4$ and 14. The modified spin-wave theory results agree very well with exact results for both sizes. The largest descrepency (less than 15%) occurs around $T = J_F$, at which independent spins start to correlate. This regime is of less interest for the purpose of this paper. In both the high temperature limit ($T > 10J_F$) and the low temperature limit ($T < \Delta_{SW}$), the modified spin-wave theory gives exact results. This is a very impressive result since the spin-wave theory is in principle developed around $T = 0$, describing the low temperature excitations of the ferromagnetic spin chain. In addition, we are able to show that boundary conditions have only weak effects on the rapidly changing susceptibility. In particular, behavior in the high temperature limit and the low temperature limit remains unchanged.

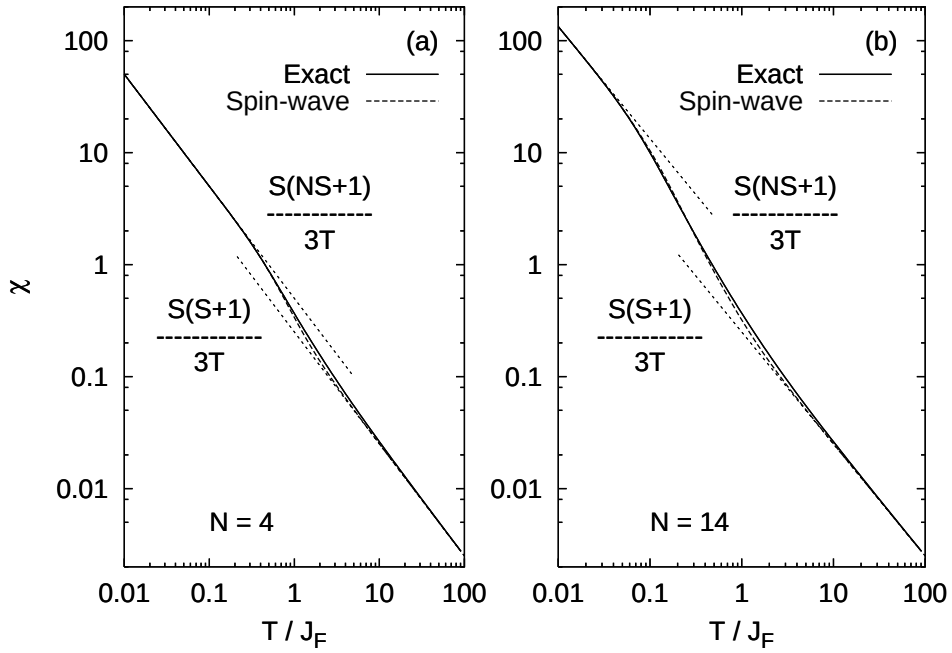

FIG. 5: Exact diagonalization result (solid line) and the modified spin-wave theory result (dashed line) of the magnetic susceptibility per spin for (a) $N = 4$ and (b) $N = 14$ ferromagnetic spin segments (S = 1/2) with periodic boundary conditions. The two dotted lines are the high-temperature Curie susceptibility of independent spins and the low-temperature Curie susceptibility of the spins when frozen into a block spin.

APPENDIX B: COUPLING BETWEEN TWO FERROMAGNETIC SPIN SEGMENTS

The problem of two ferromagnetic spin segment coupled by an antiferromagnetic bond is a special case of more general random spin chains. For small systems, we can compare our modified spin-wave theory with exact diagonalization. Figure 6 shows the susceptibility for two ferromagnetic segments ($J_F = 1$) coupled with a $J_{AF} = 0.01$ antiferromagnetic bond. The numbers of spins in the two segments are $N_1 = 6$ and $N_2 = 7$. Three regimes where the susceptibility obeys $1/T$-law are well-defined in Fig. 6. The corresponding pictures from high to low temperature are independent spins, two independent spin segments, completely locked spins, respectively. The susceptibility can be fit very well to the Curie law obtained from the simple pictures. In these Curie regimes, the modified spin-wave theory gives exact results. Discrepancies between the modified spin-wave results and exact results can only been noticed at the two crossover regimes between them. Crossover temperature scales can be roughly estimated by the smaller spin-wave gap Δ_{SW} of the two segments and the effective antiferromagnetic coupling J' between the two segments, as marked by arrows in Fig. 6.

In the RSRG scheme, one introduces an effective antiferromagnetic coupling J' between the two segments, which replaces the coupling between the two neighboring end spins. By inspecting the energy spectrum of the two-segment system in Fig. 7, one can explore the validity of the replacement. At low enough temperatures ($T < \Delta_{SW}$), the finite spin-wave excitations are frozen within each segment. Therefore, only $k = 0$ modes are relevant when the two segments couple with the effective J'. This results a group of lowest energy levels ($S = 1/2, 3/2, ..., 13/2$), which are separated from higher energy levels by a energy gap ($\sim \Delta_{SW}$). The gap is expected to become comparable with the width of the $k = 0$ energy levels, when J_{AF} is close to J_F, when RSRG argument becomes less accurate. For small J_{AF}, these $k = 0$ modes obtained from the exact diagonalization are in good agreement with the effective levels in the RG picture (the inset of Fig. 7). The difference in energy levels given by the two methods grows with increasing J_{AF}. Nevertheless, the agreement in the susceptibility obtained from the two methods still persists even when $J_{AF} \sim J_F$, as long as $T \ll \Delta_{SW}$, which poses a temperature restriction on the RSRG scheme.

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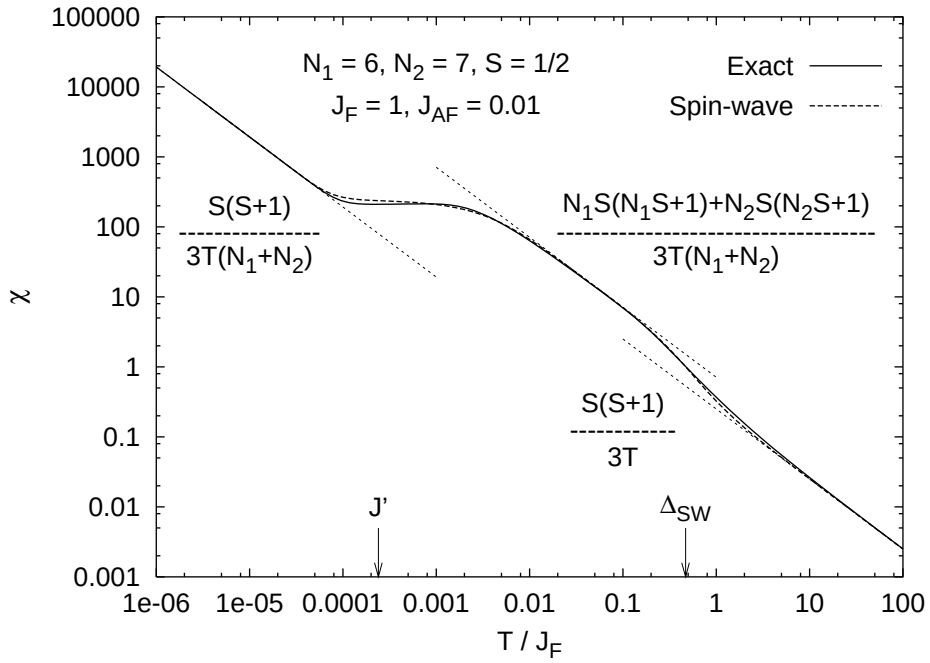

FIG. 6: Exact diagonalization result (solid line) and the modified spin-wave theory result (dashed line) of the magnetic susceptibility per spin of two spin-1/2 ferromagnetic segments ($N_1 = 6$ and $N_2 = 7$) coupled by a $J_{AF} = 0.01 J_F$ antiferromagnetic bond. The three dashed lines describe the Curie behavior in the following three regimes. (1) At high temperatures, spins are independent. (2) At intermediate temperatures, spins within each ferromagnetic segments are locked into a block spin with no internal excitations. The two block spins interacts with each other through the effective antiferromagnetic coupling (J') generated by the antiferromagnetic bond (J_{AF}). (3) At low temperatures, the two block spins are locked into one S = 1/2 spin.

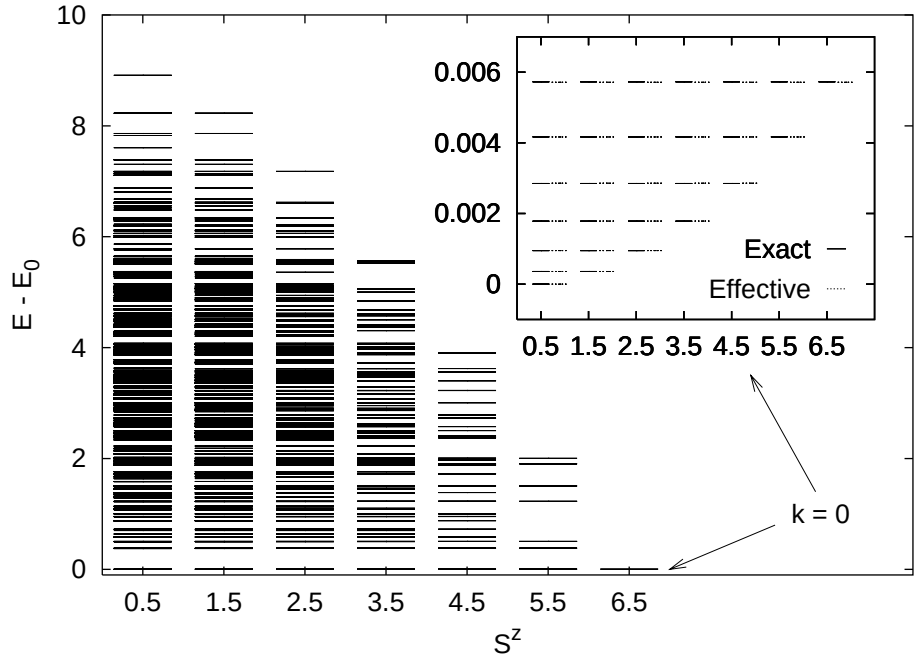

FIG. 7: Energy spectrum of the two ferromagnetic segments described in Fig. 6 ($N_1 = 6$ and $N_2 = 7$ spin segments coupled by a $J_{AF} = 0.01$ antiferromagnetic bond). The lowest energy manifold ($k = 0$ modes) of the exact diagonalization result (solid lines), shown on an amplified scale in the inset, is compared with the energy spectrum of the two block spins coupled by the effective coupling J' (Eq. 10).

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